Lyapunov instability and collective tangent space dynamics of fluids

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Abstract. The phase space trajectories of many body systems charateristic of simple fluids are highly unstable. We quantify this instability by a set of Lyapunov exponents, which are the rates of exponential divergence, or convergence, of initial (infinitesimal) perturbations along carefully selected directions in phase space. It is demonstrated that the perturbation associated with the *maximum* Lyapunov exponent is localized in space. This localization persists in the large-particle limit, regardless of the interaction potential. The perturbations belonging to the *smallest positive* exponents, however, are sensitive to the potential. For hard particles they form well-defined long-wavelength modes. The modes could not be observed for systems interacting with a soft potential due to surprisingly large fluctuations of the local time-dependent exponents.

1 Lyapunov spectra

Recently, molecular dynamics simulations have been used to study many body systems representing simple fluids or solids from the point of view of dynamical systems theory. Due to the convex dispersive surface of the atoms, the phase trajectory of such systems is highly unstable and leads to an exponential growth, or decay, of small (infinitesimal) perturbations of an initial state along specified directions in phase space. This so-called Lyapunov instability is described by a set of rate constants, the Lyapunov exponents $\{\lambda_l, l = 1, \ldots, D\}$, to which we refer as the Lyapunov spectrum. Conventionally, the exponents are taken to be ordered by size, $\lambda_l \geq \lambda_{l+1}$. There are altogether $D = 2dN$ exponents, where d and D denote the dimensions of space and of phase space, respectively, and N is the number of particles. For fluids in nonequilibrium steady states close links between the Lyapunov spectrum and macroscopic dynamical properties, such as transport coefficients, irreversible entropy production, and the Second Law of thermodynamics, have been established [1,2,3,4,5,6]. This important result provided the motivation for us to examine the spatial structure of the perturbed states associated with the various exponents. Here we present some of our results for two simple many-body systems representing dense two-dimensional fluids in thermodynamic equilibrium. The first model consists of N hard disks (HD) interacting with hard elastic collisions, the second of N soft disks interacting with a purely repulsive Weeks-Chandler-Anderson (WCA) potential.

The instantaneous state of a planar particle system is given by the $4N$-dimensional phase space vector $\boldsymbol{\Gamma} = \{\mathbf{r}_i, \mathbf{p}_i, ; i = 1, \ldots, N\}$, where $\mathbf{r}_i$ and $\mathbf{p}_i$ denote the respective position and linear momentum of molecule i. An infinitesimal perturbation $\delta\boldsymbol{\Gamma} = \{\delta\mathbf{r}_i, \delta\mathbf{p}_i; i = 1, \ldots, N\}$ evolves according to motion equations obtained by linearizing the equations of motion for $\boldsymbol{\Gamma}(t)$. For ergodic systems there exist $D = 4N$ orthonormal initial vectors $\{\delta\boldsymbol{\Gamma}_l(0); l = 1, \ldots, 4N\}$ in tangent space, such that the Lyapunov exponents

$$\lambda_l = \lim_{t \to \infty} \frac{1}{t} \ln \frac{|\delta\boldsymbol{\Gamma}_l(t)|}{|\delta\boldsymbol{\Gamma}_l(0)|} \quad , \quad l = 1, \ldots, 4N. \tag{1}$$

exist and are independent of the initial state [7,8]. Geometrically, the Lyapunov spectrum describes the stretching and contraction along linearly-independent phase space directions of an infinitesimal hypersphere co-moving with the flow. For equilibrium systems the symplectic nature of the motion equations assures the conjugate pairing rule to hold [9]: the exponents appear in pairs with a vanishing pair sum, $\lambda_l + \lambda_{4N+1-l} = 0$. Thus, only the positive half of the spectrum $\{\lambda_{1 \le l \le 2N}\}$ needs to be calculated. The sum of all Lyapunov exponents vanishes, which according to Liouville's theorem is a manifestation of the conservation of phase volume by Hamiltonian systems. Six of the exponents, $\{\lambda_{2N-2 \le l \le 2N+3}\}$, always vanish as a consequence of the conservation of energy, momentum, and center of mass, and of the non-exponential time evolution of a perturbation vector parallel to the phase flow.

For the computation of a complete spectrum a variant of the classical algorithm by Benettin *et al.* and Shimada *et al.* is commonly used [10,11]. It requires following the time evolution of the reference trajectory and of an orthonormal set of tangent vectors $\{\delta\boldsymbol{\Gamma}_l(t); l = 1, \ldots, 4N\}$, where the latter is periodically re-orthonormalized with a Gram-Schmidt (GS) procedure after consecutive time intervals Δt_{GS}. The Lyapunov exponents are determined from the time-averaged renormalization factors. For the hard disk systems the free evolution of the particles is interrupted by hard elastic collisions and a linearized collision map needs to be calculated. An algorithm based on this idea has been developed by Dellago *et al.* [12]. Although we make use of the conjugate pairing symmetry and compute only the positive branch of the spectrum, we are presently restricted to about 1000 particles by our available computer resources.

For our numerical work reduced units are used. In the case of the Weeks-Chandler-Anderson interaction potential,

$$\phi(r) = \begin{cases} 4\epsilon[(\sigma/r)^{12} - (\sigma/r)^6] + \epsilon, & r < 2^{1/6}\sigma \\ 0, & r \ge 2^{1/6}\sigma. \end{cases} \tag{2}$$

the particle mass m, the particle diameter σ, and the time $(m\sigma^2/\epsilon)^{1/2}$ are unity. In this paper we restrict our discussion to a thermodynamic state with a total energy per particle, E/N, also equal unity. For the hard-disk fluid, $(Nm\sigma^2/K)^{1/2}$ is taken as the unit of time, where K is the total kinetic energy, equal to the total energy E of the system. There is no potential energy in this case. For the reduced temperature we have $T = K/N$, where Boltzmann's constant is also

unity. In the following, Lyapunov exponents for the two model fluids will be compared for equal temperatures (and not for equal total energy). This requires a rescaling of the hard-disk exponents by a factor of $\sqrt{(K/N)_{WCA}/(K/N)_{HD}}$ to account for the difference in temperature. All our simulations are for a reduced density $\rho \equiv N/V = 0.7$, where the simulation box is a square with a volume $V = L^2$ and a side length L. Periodic boundaries are used throughout.
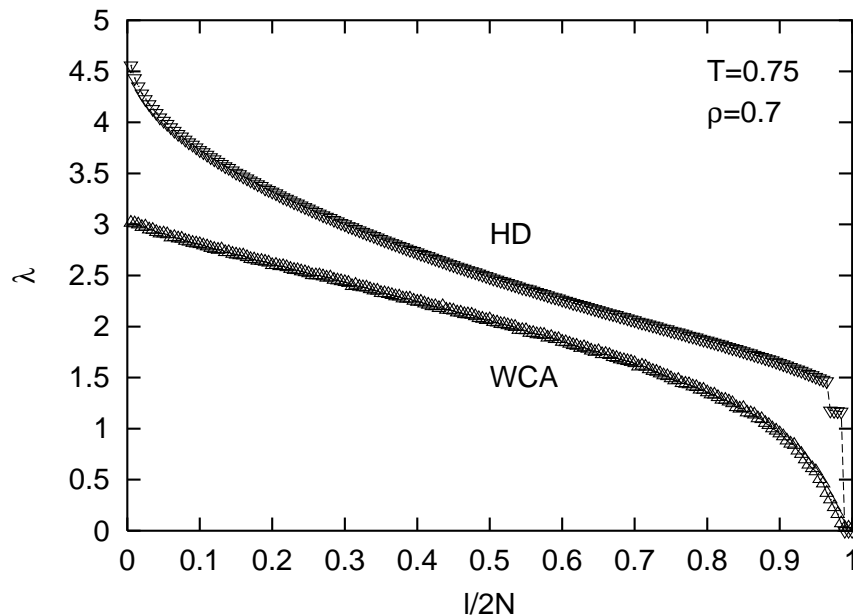
Fig. 1. Lyapunov spectrum of a dense two-dimensional fluid consisting of $N = 100$ particles at a density $\rho = 0.7$ and a temperature $T = 0.75$. The label WCA refers to a smooth Weeks-Chandler-Anderson interaction potential, and HD indicates the hard disk system.

As an example, in Fig. 1 we compare the Lyapunov spectrum of a 100-particle WCA fluid to a spectrum for an analogous hard disk system at the same temperature ($T = 0.75$) and density ($\rho = 0.7$). A reduced index $l/2N$ is used on the abscissa. It is surprising that the two spectra differ so much in shape and size. The difference persists in the thermodynamic limit. The step-like structure displayed by the hard disk spectrum for $l/2N$ close to 1 is an indication of a coherent wave-like shape of the associated perturbations, to which we refer as Lyapunov modes [13,14]. We defer the discussion of these modes to Section 4.

2 Time-dependent local exponents

We infer from Equ. (1) that the Lyapunov exponents are time averages over an (infinitely) long trajectory and, therefore, are global properties of the system. They can be rewritten as [1,15]

$$\lambda_l = \lim_{\tau \to \infty} \int_0^\tau \lambda'_l(\mathbf{\Gamma}(t)) dt \equiv \langle \lambda'_l \rangle, \tag{3}$$

where the (implicitly) time-dependent function $\lambda'_l(\mathbf{\Gamma})$ depends on the state $\mathbf{\Gamma}(t)$ which the system occupies in phase space at time t. Thus, $\lambda'_l(\mathbf{\Gamma})$ is called a local Lyapunov exponent. It may be estimated from

$$\lambda'_l(\mathbf{\Gamma}(t)) = \frac{1}{\Delta t_{GS}} \ln \frac{|\delta\mathbf{\Gamma}_l(\mathbf{\Gamma}(t + \Delta t_{GS})|}{|\delta\mathbf{\Gamma}_l(\mathbf{\Gamma}(t)|}, \tag{4}$$

where t and $t + \Delta t_{GS}$ refer to times immediately after consecutive Gram-Schmidt re-orthonormalization steps. Its time average along a trajectory is denoted by $\langle \cdots \rangle$ and gives the global exponent λ_l.
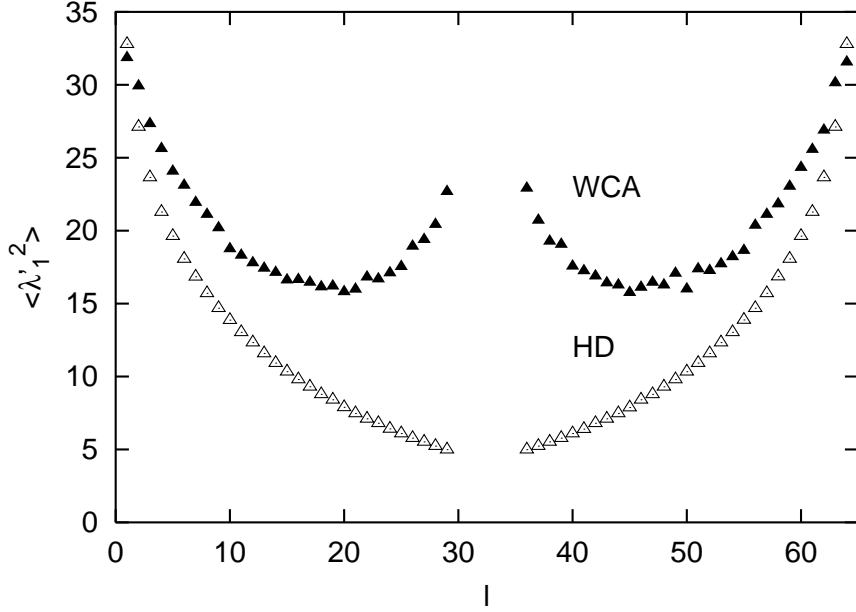
Fig. 2. Second moment of all local Lyapunov exponents for a planar 16-particle system. The labels WCA and HD refer to the Weeks-Chandler-Anderson and hard disk models, respectively. The re-orthonormalization time $\Delta t_{GS} = 0.075$, the temperature $T = 0.75$, and the density $\rho = 0.7$.

The local exponents fluctuate considerably along a trajectory. This is demonstrated in Fig. 2, where we have plotted the second momens $\langle \lambda_l'^2 \rangle$ as a function of the Lyapunov index l, $1 \leq l \leq 4N$, for a system of 16 particles, both for the WCA and HD models. $l = 1$ refers to the maximum, and $l = 64$ to the most-negative exponent. The points for $30 \leq l \leq 35$ correspond to the 6 vanishing exponents and are not shown. We infer from this figure that for the soft WCA particles the fluctuations of the local exponents become very large for the Lyapunov exponents describing relatively-weak instabilities with near-zero growth rates, $l \to 2N$. For the hard disk system, however, the relative importance of the fluctuations becomes minimal for the same exponents. We shall return to this point in Section 4.

We note that the computation of the second moments $\langle \lambda_l'^2 \rangle$ for the hard disk system requires some care. Due to the hard core collisions they depend strongly on Δt_{GS}. The variance of the fluctuating local exponents, $\langle \lambda_l'^2 \rangle - \langle \lambda_l' \rangle^2$, varies with $1/\Delta t_{GS}$ for small Δt_{GS}, as is demonstrated in Fig. 3 for $l = 1$. However, the shape of the fluctuation spectrum is hardly affected by the size of the renormalization interval.
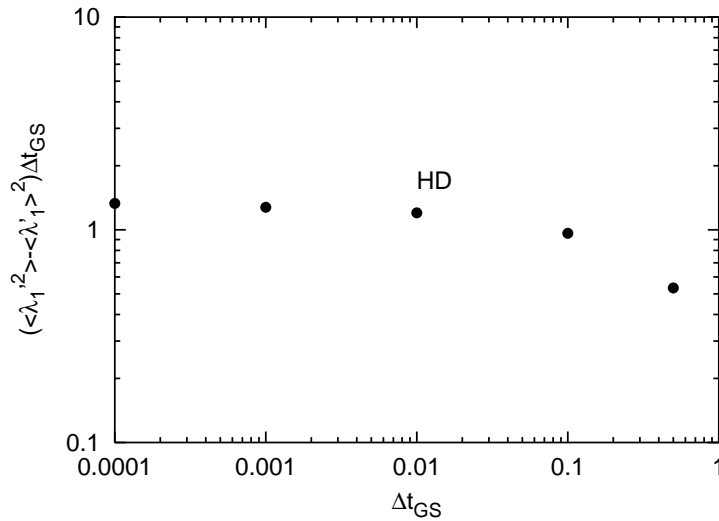
Fig. 3. Mean-square fluctuations of the local exponent λ_1', $\langle \lambda_1'^2 \rangle - \langle \lambda_1' \rangle^2$, times Δt_{GS}, as a function of the re-orthonormalization interval Δt_{GS}. The system consists of 64 hard disks. The temperature $T = K/64 = 1$, and the density $\rho = 0.7$.

3 The maximum exponent

The maximum Lyapunov exponent is the rate constant for the fastest growth of a phase space perturbation in a system. Thus, it is dominated by the fastest dynamical events in the fluid, binary collisions. There is strong numerical evidence for the existence of the thermodynamic limit $\{N, V \to \infty, N/V \text{constant}\}$ for λ_1 and, hence, for the whole spectrum [12,14]. Furthermore, the perturbation belonging to λ_1 is strongly localized in space [16,17,14]. This may be demonstrated by projecting the tangent vector $\delta\boldsymbol{\Gamma}_1$ onto the four-dimensional subspaces spanned by the perturbation components of the individual particles. The squared norm of this projection, $\delta_i^2(t) \equiv (\delta\mathbf{r}_i)_l^2 + (\delta\mathbf{p}_i)_l^2$, indicates how active a particle i is engaged in the growth process of the pertubation associated with λ_1 [18,17].
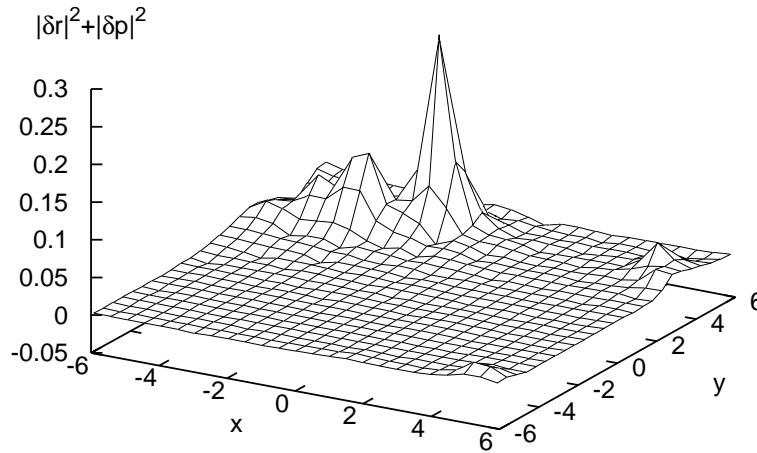
Fig. 4. The surface depicts the instantaneous contributions of the individual particles, plotted at the position of the particles in the simulation cell, to the squared norm of the perturbation vector associated with the maximum exponent λ_1. The system consists of 100 hard disks. The temperature $T = K/N = 1$, and the density $\rho = 0.7$.

In Fig. 4 $\delta_i^2(t)$ is plotted along the vertical axis for all particles of a hard disk system at the respective positions (x_i, y_i) of the disks in space, and the ensuing surface is interpolated over a periodic grid covering the simulation box.

The figure refers to an instantaneous condition for a well-relaxed system, and no averaging is involved. A strong localization of the active particles is observed at any instant of time. Similar, albeit slightly broader peaks are observed for the WCA system or other soft disk potentials.

This localization is a consequence of two mechanisms: firstly, after a collision the delta-vector components of two colliding molecules are linear functions of their pre-collision values and have only a chance of further growth if their values before the collision were already far above average. Secondly, each renormalization step tends to reduce the (already small) components of the other non-colliding particles even further. Thus, the competition for maximum growth of tangent vector components favors the collision pair with the largest components. The active zone moves in space in a diffusive manner.

The localization also persists in the thermodynamic limit. To show this we follow Milanović *et al.* [14] and order all squared components $[\delta\boldsymbol{\Gamma}_1]_j^2; j = 1, \ldots, 4N$ of the perturbation vector $\delta\boldsymbol{\Gamma}_1$ according to size. By adding them up, starting with the largest, we determine the smallest number A of terms, required for the sum to exceed a threshold Θ. Then, $C_{1,\Theta} \equiv A/4N$ may be taken as a relative measure for the number of components actively contributing to λ_1:

$$\Theta \leq \left\langle \sum_{s=1}^{4NC_{1,\Theta}} [\delta\boldsymbol{\Gamma}_1]_s^2 \right\rangle, \qquad [\delta\boldsymbol{\Gamma}_1]_i^2 \geq [\delta\boldsymbol{\Gamma}_1]_j^2 \text{ for } i < j. \tag{5}$$

Obviously, $C_{1,1} = 1$.

In Fig. 5 $C_{1,\Theta}$ is shown for $\Theta = 0.90$ as a function of the particle number N, both for the WCA fluid and for the hard disk system. It converges to zero if our data are extrapolated to the thermodynamic limit, $N \to \infty$. This supports our assertion that in an infinite system only a *vanishing* part of the tangent-vector components (and, hence, of the particles) contributes significantly to the maximum Lyapunov exponent at any instant of time.
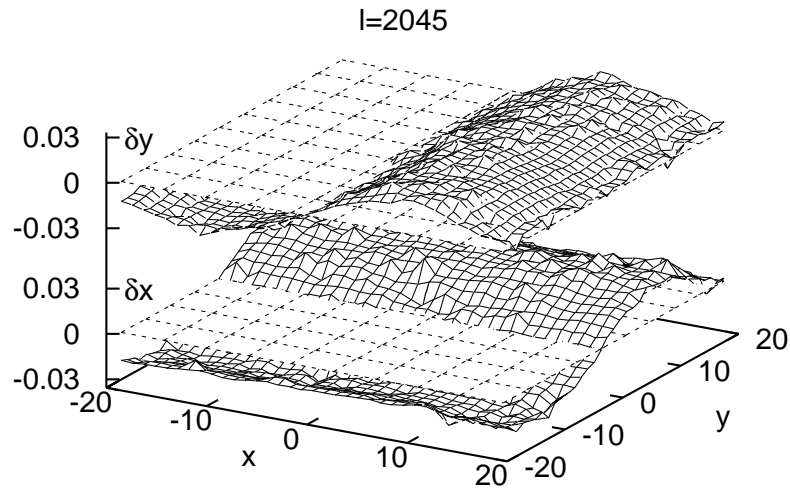
4 Lyapunov modes

We have already mentioned the appearance of a step-like structure in the Lyapunov sepctrum of the hard disk system for the positive exponents closest to zero, which was first observed in Ref. [12]. They are a consequence of coherent wave-like spatial patterns generated by the perturbation vector components associated with the individual particles [13,14].
In Fig. 6 this is visualized by plotting the perturbations in the x (bottom surface) and y directions (top surface), $\{\delta x_i, i = 1, \ldots, N\}$ and $\{\delta y_i, i = 1, \ldots, N\}$, respectively, along the vertical axis at the instantaneous particle positions (x_i, y_i) of all particles i. This figure depicts a transverse Lyapunov mode of type T_1, for which the perturbation components are perpendicular to the respective wave vectors with one wave length equal to L, the linear extension of the simulation box. The system consists of $N = 1024$ hard disks, and the perturbation vector considered in the figure belongs to the smallest positive exponent λ_{2045}. An
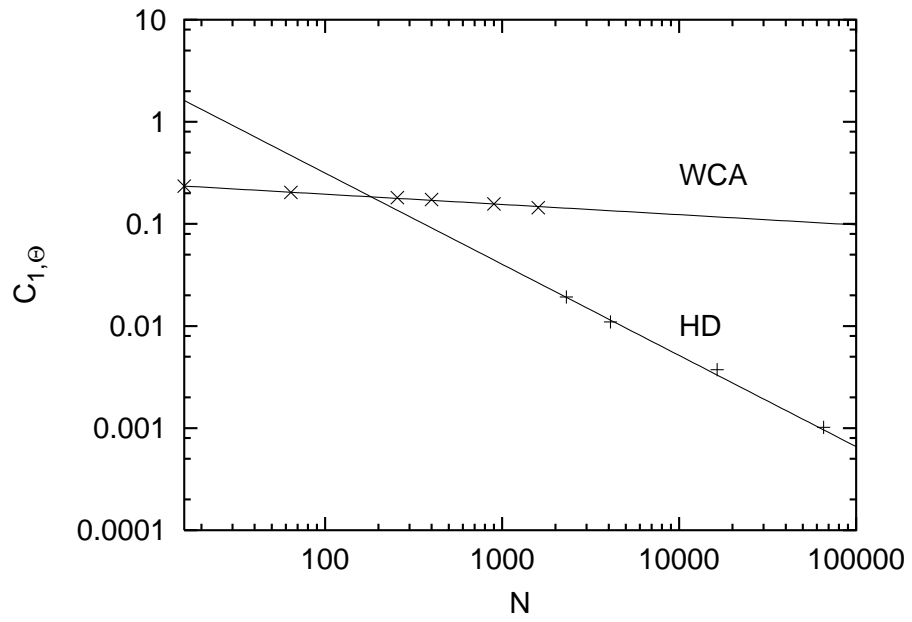
Fig. 5. Dependence of the localization measure, $C_{1,\Theta}$, on the number of particles N for the parturbation associated with the maximum Lyapunov exponent λ_1. The threshold $\Theta = 0.90$. The soft and hard disk results are labeled by WCA and HD, respectively.

Fig. 6. Transverse Lyapunov mode of type T_1 associated with the smallest positive exponent λ_{2045} for 1024 hard disks at a density $\rho = 0.7$. The temperature $K/N = 1$.

Bottom: Plot of the tangent vector components δx of the individual particles (along the vertical axis) for all particles at their instantaneous postions (x, y) in the simulation box (horizontal plane). Top: Analogous plot for δy.

analogous plot of δp_x and δp_y for the same perturbation vector is identical in shape to that of δx and δy in Fig. 6, with the same phases of the waves. This is a consequence of the fact that the perturbations are solutions of linear first-order equations instead of second order equations such as the wave equation. Furthermore, the exponents for $2042 \leq l \leq 2045$ are equal. This four-fold degeneracy of non-propagating transversal modes, and an analogous eight-fold degeneracy of propagating longitudinal modes, are responsible for a complicated step structure for l close to $2N$, which has been studied in detail in Refs. [13,19].

The wave length of the modes and the value of the corresponding exponents are determined by the size L of the square simulation box. There is a kind of linear dispersion relation [13,14] indicating that the smallest positive exponent is proportional to $1/L$. This assures that for a square simulation box with aspect ratio 1 there is no positive lower bound for the positive exponents of a hard disk system in the thermodynamic limit [14,19].

So far, our discussion of modes applies only to hard disk fluids. In spite of a considerable computational effort we have not yet been able to indentify modes for two-dimensional fluid systems with a soft interaction potential such as WCA or similar potentials [20]. This surprising result seems to be due to the very strong fluctuations of the local exponents discussed in Section 2. They tend to obscure any mode in the system in spite of considerable averaging and make a positive identification very difficult.

References

1. H. A. Posch and W. G. Hoover, Phys. Rev. A **38**, 473 (1988).
2. H. A. Posch and W. G. Hoover, Phys. Rev. A **39**, 2175 (1989).
3. D. J. Evans and G. P. Morriss, *Statistical mechanics of nonequilibrium liquids*, Academic Press, London, 1990.
4. Wm. G. Hoover, *Computational Statistical Mechanics*, (Elsevier, New York, 1991); *Time Reversibility, Computer Simulation, and Chaos*, (World Scientific, Singapore, 1999).
5. P. Gaspard, *Chaos, Scattering, and Statistical Mechanics*, (Cambridge University Press, 1998).
6. J. R. Dorfman, *An Introduction to Chaos in Nonequilibrium Statistical Mechanics*, (Cambridge University Press, 1999).
7. V. I. Oseledec, Trans. Mosc. Math. Soc. **19**, 197 (1968).
8. J.-P. Eckmann and D. Ruelle, Rev. Mod. Phys. **57**, 617 (1985).
9. D. Ruelle, J. Stat. Phys. **95**, 393 (1999).
10. G. Benettin, L. Galgani, A. Giorgilli, and J. M. Strelcyn, Meccanica **15**, 9 (1980).
11. I. Shimada and T. Nagashima, Proc. Theor. Phys. **61**, 1605 (1979).
12. Ch. Dellago, H.A. Posch, and Wm. G. Hoover, Phys. Rev. E **53**, 1485 (1996).
13. H. A. Posch and R. Hirschl, "Simulation of Billiards and of Hard-Body Fluids", in *Hard Ball Systems and the Lorenz Gas*, edited by D. Szasz, Encyclopedia of the mathematical sciences **101**, p. 269, Springer Verlag, Berlin (2000).

14. Lj. Milanović and H. A. Posch, Localized and delocalized modes in the tangent-space dynamcs of planar hard dumbbell fluids, J. Molec. Liquids (2002), in press.

15. Wm. G. Hoover, Ch. Dellago, and H. A. Posch, Fluctuations, convergence times, correlation functions, and power laws from many-body Lyapunov spectra for soft and hard disks and spheres, submitted.

16. Wm. G. Hoover, K. Boercker, and H. A. Posch, Phys. Rev. E **57**, 3911 (1998).

17. Lj. Milanović, H .A. Posch, and Wm. G.Hoover, Molec. Phys., **95**, 281 - 287 (1998).

18. Lj. Milanović, H. A. Posch, and Wm. G. Hoover, Chaos, **8**, 455 (1998).

19. Ch. Forster, Lj. Milanović, and H. A. Posch, in preparation.

20. Wm. G. Hoover, H. A. Posch, Ch. Forster, Ch. Dellago, and M. Zhou, Lyapunov modes of two-dimensional many-body systems; soft disks, hard disks, and rotors, J. Stat. Phys., submitted.